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Synthesis of Iron-ferrocyanide functionalized magnetic nanocluster for the removal of cesium

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1. Introduction

Magnetic nanoadsorbents composed of a magnetic particles core and functional shell, which adsorb the contaminants, has attracted significant attention in environmental remediation owing to their high surface area and unique superparamagnetism.[1] The nuclear accident at the Fukushima Daiichi nuclear power station in 2011 released a huge quantity of radioactive contaminants into the environment. Among these, cesium Cs-137 is the most problematic contaminant due to its long half-life (30.2 years), and high-energy gamma ray (γ-ray) emissions.[2] Among various adsorbents to treat Cs-137 contaminated water, metal ferrocyanides were widely applied to remove the Cs-137 in water.[3] For better separation of metal ferrocyanide from water, recently, our group reported the fabrication of copper ferrocyanide-functionalized magnetic nanoparticles (Cu-FC-EDA-MNPs) using alkoxysilanes, having ethylenediamine (EDA) group, modified Fe_3O_4 nanoparticles (EDA-MNPs) for the fast and easy magnetic separation of metal ferrocyanide.[4] However, the fabrication method was multistep procedure. Thus, a more simplified fabrication procedure is still desired.

In the present study, magnetite nanocluster was synthesized by hydrothermal method, and coated with iron ferrocyanide for the adsorption of cesium in an aqueous solution through simple addition of iron ferrocyanide in acid condition. We describe the morphology, structure, and physical property of these nanoparticles. In addition, their ability to eliminate cesium from water was also evaluated

2. Methods

Magnetic nanocluster were synthesized through hydrothermal reaction. Iron chloride were firsit dissolved in eghylene glycol under vigorous stirring. A mixture solution was obtained after stirring for one hour. Then, sodium acetate were added to this solution. After storring for another 30 minute, the resultant solution was transferred into a Teflon-lined stainless-steel autoclave. The autoclave was sealved and heated at 200°C for some hours, and cooled to room temperature. The resultant were collected with the help of magnet, followed by washing with deioned water for several times, then the product was dried in vacuum at 60°C for overnight.

The dried magnetic nanoclusted was added into the potassium iron ferrocyanide solution within the acid condition. Then, the mixture was stirred for several hours. The iron ferrocytanide founctionalized magnetic nanocluster was collected by magnet, then washed with water several time. Finally, the product was dried in an oven at 60°C for overnighte.

3. Results

The magnetic nanocluster (MNC) was synthesized by hydrothermal reaction in the polytol ethyleneglycol at elevated temperature. An X-ray diffraction (XRD) analysis confirmed the crystalline nature of the MNC. All peaks agreed well with the values reported previously for the magnetite (Fe_3O_4).

Fig. 1(a) shows the transmittance electron microscopy (TEM) image of MNC. From the TEM image, the nanosized nanoparticels were aggregated during the hydrothermal reaction. The average size of MNC obtained from the TEM images was in the range from 150 nm to 250 nm.

In the next step, magnetic nanocluster (MNC) was further functionalized with Iron and ferrocyanide. It is known that potassium iron ferrocyanide can easily form the iron forrocyanide that can absorb the cesium in the presence of iron ions and acid condition. The magnetic nanocluster works as a source of iron ions and template for the iron ferrocyande crystals.

The FTIR spectra of the iron ferrocyanide (IFC)-MNC also showed the successful immobilization of ferrocyanide (Fig. 2). A strong absorption peak of cyanide groups appears at 2050 cm^{-1} in IFC-MNC. Therefore, it was concluded that the surface of the magnetite nanocluster was successfully functionalized with iron ferrocyanide.)). As the added amount of ferrocyanide was increased, the intensity of absorption peak of cyanide groups of IFC-MNPs was also increased, indicating that larger amount of iron ferrocyanide crystal was formed in the surface of magnetic nanocluster.

Fig. 1(b) shows the transmittance electron microscopy (TEM) image of Iron ferrocyanide functionalized MNC (IFC-MNC). From the TEM image, the nanosized nanoparticels were still aggregated during the coating procedure of iron ferrocyanide and the MNC were coated with other materials, iron ferrocyanide. This result indicates that the coating procedure didn't change the MNC morphology. The average size of IFC-

MNC obtained from the TEM images was in the range from 200 nm to 350 nm.

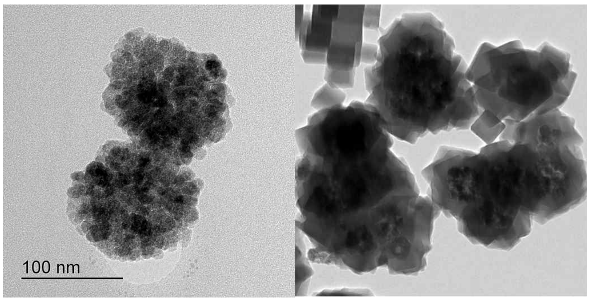

Fig. 1. TEM images of (a) magnetic nanocluster and (b) iron ferrocyanide functionalized magnetic nanocluster in aqueous solution.

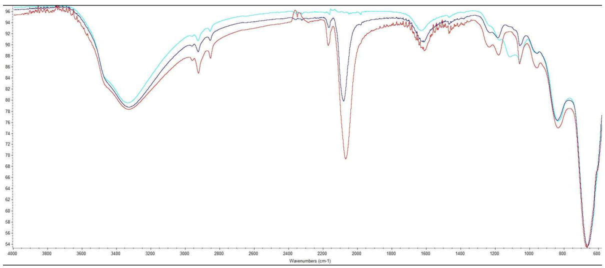

Fig. 2.FTIR spectra of magnetic nanocluster (sky blue) and iron ferrocyanide functionalized magnetic nanocluster (red & blue).

Next, the Cs removal efficiency of the IFC-MNC was examined. Various concentrations of the IFC-MNC were mixed with an aqueous solution containing Cs. The IFC-MNC exhibited good removal efficiency even at lower concentrations.

4. Conclusions

In this study, we fabricated Iron ferrocyanide immobilized magnetite nanocluster (IFC-MNC) using hydrothermal methods. The CIFC-MNC exhibited easy separation ability from water by an external magnet, and showed a high removal efficiency of cesium in aqueous solutions. Therefore, the IFC-MNC demonstrated good potential for the treatment of water contaminated with radioactive cesium.

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